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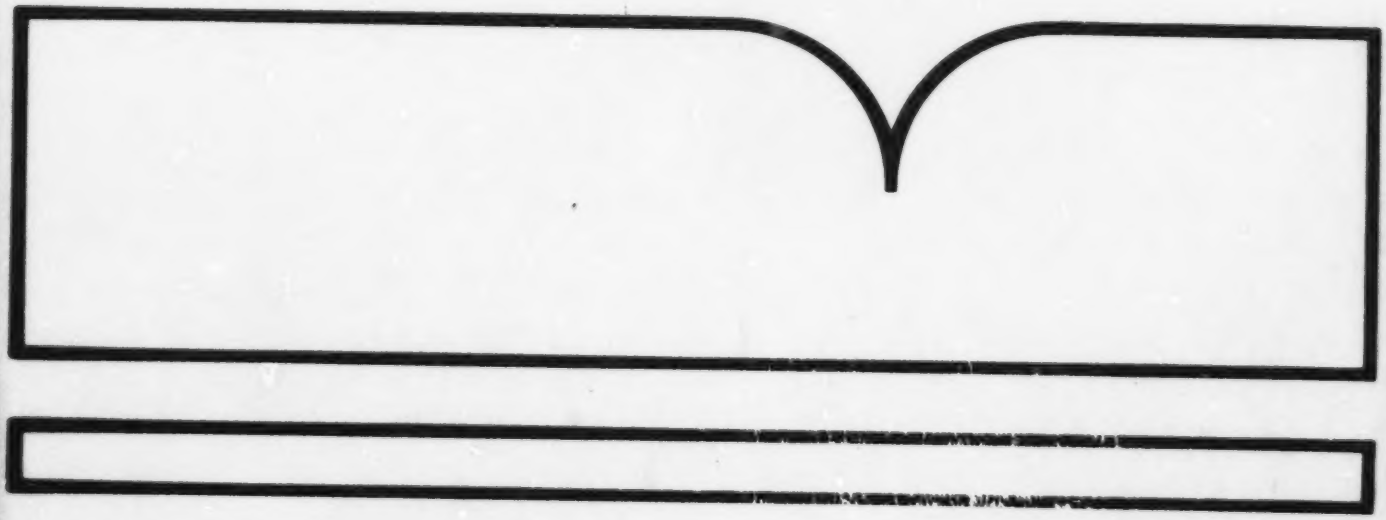

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A Multivariate Assessment of Meteorological Influences on Inhalable Particle Source Impacts

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ABSTRACT

This paper identifies the sources of fine and coarse inhalable particles at a site in metropolitan Boston and investigates their respective relationships to meteorological conditions. In this work, Principal Component Analysis (PCA) is applied to: 1) fine particle mass elemental data; 2) coarse particle mass elemental data, and; 3) meteorological measurements (primarily collected at nearby Logan International Airport). In addition to local surface observations, air mass trajectory information concerning each sampling day is included in the meteorological data set, allowing the consideration of air mass transport as one factor in particle impacts. As part of these PCA analyses, four different objective component selection criteria are examined and compared. The Scree test of eigenvalues is found to result in the most interpretable components for the specific air pollution and meteorological data sets considered in this work.

The rotated PCA analyses result in the identification of five fine mass particle source classes (soil, motor vehicle, coal related, oil and salt aerosols); six coarse mass particle source classes (soil, motor vehicle, refuse incineration, residual oil, salt and coarse sulfate aerosols); and five meteorological components (air mass transport over the eastern seaboard to Boston, moisture influences, poor local dispersion, season and air mass transport over the Ohio Valley/Midwest to Boston). It is found that local combustion sources (e.g., residual oil burning and motor vehicles) recorded their highest impacts during poor local dispersion and mesoscale advection from downtown Boston. Coarse soil impacts are highest at this site during dry, summertime conditions (when fugitive dust generation is at a maximum). Coal combustion-related fine particle impacts, however, are at a maximum during air mass transport into the Boston vicinity (especially from the Ohio Valley/Midwest direction), regardless of local dispersion conditions. It became obvious that meteorology plays an important role in the extent to which these particle source classes impact the monitoring site on a given day, and that it is, in large measure, the highly variable weather conditions in Boston which allow statistical approaches to discriminate the various source classes. Based on these analyses, it is concluded that both local and transported aerosols have an influence on inhalable aerosol exposures at this site, with the sulfate portion being most correlated with polluted air mass transport into the Boston area.

1. Introduction

Investigations into the sources of particulate pollution have employed a wide range of statistical models. Gaussian plume source models, for example, are routinely applied in conjunction with meteorological and estimated pollutant emission data. Such models make assumptions regarding pollutant plume behavior to predict the downwind impacts of point and area pollution sources (e.g., Turner, 1970). This allows an estimation of contributions by specific sources under given meteorological conditions. Conversely, receptor oriented, source apportionment models examine the chemical and/or physical characteristics of a collected aerosol (e.g., its elemental composition) in order to identify contributions by various sources, based upon known characteristics of those sources (e.g., elements uniquely contained in particles from a source). One such source apportionment model, the Principal Component Analysis (PCA) of particle elemental composition data, is employed in this investigation of the meteorological influences on particle sources impacting a site in metropolitan Boston, Massachusetts.

The application of PCA and other related multivariate techniques such as Factor Analysis (FA) have been shown to be useful aids in the interpretation of diverse meteorological data. Kutzbach (1967), and Walsh and Richman (1981), for example, have employed PCA analyses to derive independent weather predictors or descriptors. Both studies considered spatial fields of a single meteorological variable (e.g., sea level pressure or surface temperature fields), using PCA to derive components embodying basic weather patterns. Christensen and Bryson (1966), in contrast, used PCA to show that many intercorrelated weather variables at an individual site may be expressed by a smaller number of independent components which correspond with known meteorological regimes. More recently, Henry

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and Hidy (1979, 1982), Thurston and Spengler (1980), and Lioy *et al.* (1982) have all applied PCA to both air quality and meteorological variables in order to explain ambient sulfate concentration variations at a site over time. However, such meteorological components have not yet been applied to explain variations in particle source impacts.

Multivariate statistical methods have also been applied to arrays of pollution variables at particular sites over time and to spatial pollutant distributions during specific time intervals. The application of PCA or FA to pollutant data determines components or factors which explain a large portion of the variation in the data and which may represent the influence of individual pollutant source classes. Blifford and Meeker (1967), for example, applied rotated FA to both particle elemental composition and mass data from 30 United States cities, thereby identifying independent components of composition variance as specific particle source classes. Hopke *et al.* (1976) applied FA to Boston urban aerosol elemental compositions to identify particle contributors in that city. The components identified through such multivariate techniques may be due to fluctuations in either the rate of source emission or the dispersive effects of meteorological conditions. Henry (1977) recognized this fact and conducted an analysis of elemental mass fractions (instead of concentrations) in order to reduce the covariation caused by any general rise and fall of all elemental concentrations as local dispersion conditions varied. However, we are not interested here in the control of meteorological influences on our data, but in the elucidation of their respective roles in determining the occurrence of particle source impacts.

In this work, PCA is applied separately to: 1) two simultaneously collected fine and coarse inhalable particle elemental composition data sets; and 2) a set of meteorological data collected concurrent to particle samples. The component loadings resulting from the former PCAs are interpreted as the fine and coarse particle sources affecting the particulate matter concentrations at a monitor, while the latter PCA yields components interpretable as the underlying meteorological regimes affecting the site. The elemental (source) components considered here have previously been identified and quantified (Thurston and Spengler, 1985), and the emphasis of this paper will be to investigate the respective roles of meteorological influences on these source impacts. To achieve this, the source and meteorological components are tested for correlation with each other in order to allow an assessment of the role of each meteorological component on source impact variability. These analyses allow insight into the relative importance of different meteorological influences to each identified particle source class, while testing our prior interpretations of the source components identified during the PCA of the fine and coarse particle elemental data sets.

2. Methodology

a. Particle mass and elemental data collection

The fine and coarse inhalable particle (IP) samples examined in this work were collected using a Beckman virtual impactor dichotomous sampler (Cat. No. 675900). Fine fraction aerosols collected had an aerodynamic diameter (d_a) less than 2.5 μm, while coarse particles had 2.5 $\mu m < d_a < 15$ μm. These samples were analyzed for elements by X-ray fluorescence (XRF) spectrometry (Dzubay and Stevens, 1975). Fine and coarse masses were determined by Beta-gauge (Jaklevic, *et al.*, 1981). The 332 fine and coarse samples analyzed here were 24 h samples collected at least every other day between 4 June 1979 and 5 June 1981. The Watertown sampling site is situated in a high school athletic field located in a suburban neighborhood. The site is within the Boston metropolitan area, approximately 10 km west-northwest of downtown. This particle sampling was conducted as part of the Harvard School of Public Health's ongoing prospective epidemiologic Air Pollution Respiratory Health Study (Ferris *et al.*, 1979). The health study particle sampling network and analysis techniques have been described in detail previously (Spengler and Thurston, 1983).

The elements selected for analysis, their respective mean concentrations, standard errors of the mean (SE) and mean standard deviations (SD) of measurement uncertainty are presented, by particle size fraction, in Table 1. Prior to statistical analysis of the elemental and mass data, the samples were checked for validity. Elements thought to be unreliably determined by XRF at this site were also eliminated from the data set. This determination was made on each size fraction independently, and was based upon a qualitative knowledge of XRF elemental interference problems and a quantitative determination of the significance of elemental concentrations relative to their reported uncertainties. Further details regarding the quality assurance and data validation procedures employed may be found in Spengler and Thurston (1983) and Thurston and Spengler (1985).

b. Meteorological data acquisition

Meteorological data concurrent to 237 of the 332 IP samples were derived primarily from surface observations taken every three hours at the nearest National Weather Service station, Logan International Airport in Boston (approximately 10 km east of the Watertown site). The meteorological variables employed and their means and standard errors are presented in Table 2. The mean values presented in Table 2 compare favorably with the climatological means reported for the Logan station (NOAA, 1981). A daytime solar flux index was crudely estimated by assuming the reduction from a maximum of 1.0 to be dependent upon noontime solar elevation and inversely proportional to the

TABLE 1. Mean Watertown, MA fine and coarse aerosol elemental and mass concentrations (ng m^{-3}).

	Fine aerosol (m = 332)			Coarse aerosol (m = 332)		
Element	Mean	Standard error of mean	Mean standard deviation of measurement uncertainty	Mean	Standard error of mean	Mean standard deviation of measurement uncertainty
Al	*	*	17.1	366	25.6	51.7
Si	104	6.96	13.7	1021	66.2	158
S	1840	74.6	108	201	9.53	120
Cl	86.2	11.1	7.12	306	45.8	66.2
K	76.2	2.74	3.94	112	6.62	10.3
Ca	41.9	2.29	2.31	214	15.2	13.2
Ti	*	*	4.29	25.9	2.08	5.22
V	22.1	1.10	2.89	3.34	0.33	2.46
Mn	3.61	0.17	1.14	5.81	0.403	1.29
Fe	74.7	3.87	4.15	281	17.1	15.5
Ni	8.57	0.39	0.86	2.44	0.13	0.66
Cu	16.1	1.01	1.40	10.4	0.69	1.28
Zn	26.5	1.05	1.71	12.2	0.64	1.30
Se	0.595	0.046	0.273	*	*	0.208
Br	85.7	4.39	4.18	25.7	1.55	1.94
Pb	326	13.2	15.6	75.6	4.57	5.95
Mass ($\mu g\ m^{-3}$)	17.4	0.52	1.60	8.57	0.45	1.60

* Mean concentration below detection (1.65 × SD of measurement uncertainty).

recorded afternoon cloud cover. Precipitation index was a dimensionless measure of the rate of precipitation at each measurement hour, ranging from 0 (none) to 3 (heavy liquid precipitation). Sea level pressure tendency was defined as the midnight to midnight 24-hour change in pressure during the sampling period. Morning and afternoon ventilation factors [= (mixing height) × (layer mean wind speed)] were calculated using Logan Airport surface observations and Portland, Maine upper air soundings on the individual sampling days. Day and night stability classes were assigned as 1 through 6 (ranging from very unstable to very stable, with 4 = neutral) as estimated from Logan surface observations of wind speed and cloud cover according to the Pasquill-Gifford scheme (Turner, 1970). Unless otherwise designated, all meteorological values are 24-

TABLE 2. Means and standard errors of meteorological variables employed in PCA (m = 237).

Variable	Interpretation (units)	Mean	Standard error
FLUX	Afternoon solar flux index (dimensionless)	0.32	0.02
PRECIP	Liquid precipitation index (dimensionless)	0.08	0.01
SLPRESS	Mean sea level pressure (mb)	1015	0.4
PTEND	Sea level pressure tendency (mb day^{-1})	0.60	0.52
ATEMP	Mean ambient temperature (°F)	54	1
AVGWSPD	Mean windspeed (m s^{-1})	4.8	0.1
MVENTF	Morning mixing layer ventilation ($m^2\ sec^{-1}$)	6900	410
AVENTF	Afternoon mixing layer ventilation ($m^2\ sec^{-1}$)	10400	510
RELHM	Mean relative humidity (%)	65	1
ABSHM	Mean absolute humidity (gm $H_2O\ m^{-3}$)	8.2	0.3
NSTBCL	Nighttime stability class (dimensionless)	4.3	0.04
DSTBCL	Daytime stability class (dimensionless)	3.8	0.03
PERNE	Northeast winds (%)	5.2	0.8
PEREE	East winds (%)	6.2	0.8
PERSE	Southeast winds (%)	8.5	0.9
PERSS	South winds (%)	12.4	1.2
PERSW	Southwest winds (%)	18.3	1.4
PERWW	West winds (%)	17	1
PERNW	Northwest winds (%)	23	2
PERCES	Eastern seaboard air mass sector (%)	22	2
PERCOV	Ohio Valley/Midwest air mass sector (%)	29	2
PERCEC	Eastern Canada air mass sector (%)	26	3

h averages for periods coinciding with the particle sampling.

Since the sources of air pollution are of primary interest in this analysis, emphasis has been given to the analysis of winds during the sampling periods. Four wind trajectories of air mass origin were calculated per sampling day (one every six hours) and individually assigned to one of the five sectors displayed in Fig. 1. The trajectories were generated using the Air Resources Laboratories' Atmospheric Transport and Dispersion Model (ARL-ATAD) (Heffter, 1980). This model uses vertically averaged transport winds (from the surface to the top of the mixing layer) as recorded throughout North America to calculate trajectories. Trajectories were calculated for periods up to 72 h, moving backwards in time from Watertown, Massachusetts. These trajectories estimate the movement of the center of mass of the mixed layer of the atmosphere, so that it is possible for sources located laterally from a trajectory line to also contribute to a particular sample (Samson, 1980). Such trajectories are known to be subject to increasing errors with distance, so that errors on the order of 100 km for a 72-h back trajectory are not unexpected (e.g., see Pack *et al.*, 1978). However, trajectory errors of this order would not be expected to significantly affect the regional classifications employed here.

Wind trajectories were obtained for all days between 4 June 1979 and 18 December 1980. However, since sampling was conducted only every other day, the analysis was limited to 237 sampling days. For each sampling day for which trajectories were available, the number assigned to each sector was divided by the total number for the day (usually four) and multiplied by 100 to give the percent of trajectories from each sector during a day. Since the Heffter model relies on upper air wind soundings conducted throughout continental North America, the model underrepresents winds from the Atlantic Ocean, where no wind data are available. In part because of this, only the three most prevalent continental sectors were included in the analysis. It should be noted, however, that the Eastern Seaboard sector may yet be underrepresented if coastal trajectories passed far out over the Atlantic before reaching Boston. Local surface wind observations have also been utilized to calculate the percent occurrence of each direction during the sampling periods. However, the percent north winds have been excluded from the analysis in order to avoid a singular correlation matrix in the PCA (i.e., since the wind percentages always sum to 100%, the inclusion of all wind directions would result in an overdetermined matrix). Thus, both local and regional air mass transport wind information have been incorporated in this analysis of meteorological regimes influencing the Watertown site during particle sample collection.

c. *Statistical methods*

Principal Component Analysis is a statistical technique that assumes that variations in observable variables (e.g., elemental concentrations or meteorological measurements) are merely manifestations of the influences of a limited number of "true" underlying variables. Thus, PCA is a technique that replaces a large number of intercorrelated variables with a smaller number of independent variables. A major difference between PCA and FA is that, while components are derived by PCA to explain as much of the total dataset variance as possible, factors are derived by FA to ex-

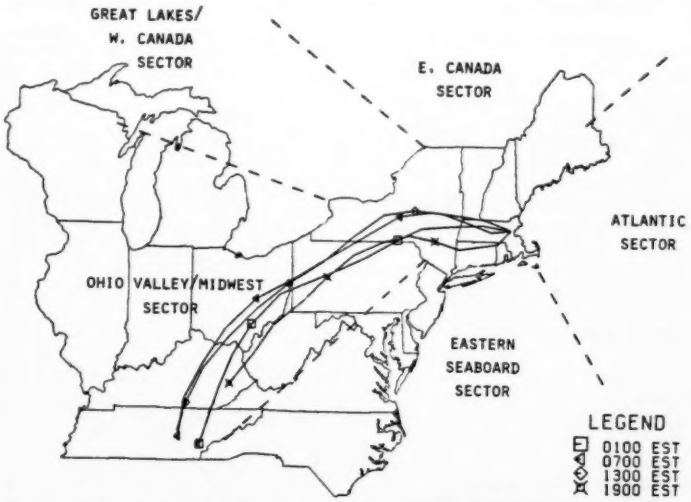

FIG. 1. Map displaying wind trajectory sector classes, with four trajectories for an example day (22 October 1979).

plain only the variance shared by the variables considered (i.e., a subset of the total variance). In the PCA model, each observable variable is assumed to be a product of the interactions of the various principal components (PCs). Based on their mathematical relationships with the observable variables, the PCs can often be identified as underlying influences on the data (e.g., pollution sources or meteorological regimes).

The first step in common PCA is the transformation of the observable data into dimensionless standardized observations:

$$Z_{ik} = \frac{(X_{ik} - \bar{X}_i)}{\sigma_i}, \qquad (1)$$

where $i = 1, 2, \cdots n$, the total number of variables in the analysis; $k = 1, 2, \cdots m$ the total number of observations; Z_{ik} (often termed the Z-score) is the standardized value of variable i for observation k; X_{ik} is the value of that variable for that observation; $\bar{X}_i$ is the mean value for the ith variable over all observations; and σ_i is the standard deviation about the mean of variable i. This, and all other statistical analyses presented herein, employed the Statistical Analysis System (SAS) (SAS Institute, 1982). If this standardization were not conducted, there would be a tendency to deemphasize those variables with smaller absolute magnitudes during the subsequent PCA. Hence, standardizing the data prior to the PCA tends to equalize the opportunity of both large and small magnitude variables to influence the analysis.

Mathematically, PCA assumes that each variable is made up of the sum of contributions from each of j underlying components of variation. Hence,

$$Z_{ik} = \sum_{j=1}^{p} W_{ij} P_{jk}, \qquad (2)$$

where P_{jk} is the j component's value for observation k; $j = 1, 2, \cdots p$, the number of underlying components influencing the data; and W_{ij} is the loading matrix of the components (i.e., the scoring coefficients). The specifics of PC derivation are presented in detail in statistics texts such as Harris (1975), Rummel (1970), and Harman (1976).

The primary objective of applying PCA to the previously described independent data sets is to derive a limited number of components for each data set which explain a majority of the total variance in the original variables. In order for the PCA reduction in dimensionality to be useful, the new variables (PCs) must have simple substantive interpretations. However, it is found empirically that several different underlying sources of variation may be incorporated into any one unrotated PC. Thus, the components resulting from unrotated PCA frequently do not have straightforward or unique interpretations (Harris, 1975). For this reason, PCA often includes the subsequent rotation of a limited number of the PCs (using a maximization criteria such as Varimax), resulting in components which have been found to be more representative of individual underlying sources of variation (e.g., Walsh and Richman, 1981). This, in turn, results in more interpretable and useful PCs. It should be noted that PCs with smaller eigenvalues (which, presumably, are dominated by error variance) are dropped before rotation. Thus the rotated component and rotated factor solutions for a data set are functionally similar in many cases. The merits of PCA rotation in analyzing air pollution data have been discussed in the literature by Thurston (1981), Henry and Hidy (1981), Hopke (1982), and Richman (1983).

Many methods are available for the rotation of PCs. These may be classed into either: 1) orthogonal rotations, where the components remain uncorrelated with one another, or 2) oblique rotations, where PCs are allowed to be intercorrelated. Each rotation class has its own advantages and disadvantages in addressing multivariate problems. The scores of orthogonally rotated PCs may be employed in subsequent multivariate regression or correlation analyses without concern that multicollinearities might confound the results, since they are, by definition, uncorrelated. Oblique component scores, conversely, are not orthogonal and can result in confounding when used in subsequent regression analyses. However, oblique rotations may yield more readily interpretable components in cases where independent PCs cannot. Thus, the use of oblique rotations provides components which may have clearer interpretations and can also provide information as to the intercorrelation of these components, but the components provided may be less appropriate for use in regression analyses than orthogonal PCA rotations. In this and a related paper (Thurston and Spengler, 1985), we employ the PC scores in subsequent statistical analyses, and orthogonal components are preferable. Based upon these considerations, orthogonal Varimax rotated PCA has been employed in these analyses of the individual elemental and meteorological data sets.

Numerous statistical techniques have been proposed to guide the "proper" number of unrotated components for retention in the rotation procedure. However, it is often the case that these criteria give conflicting guidance for a given data set (e.g., Hakstian *et al.*, 1982). As a result, the number of components retained for analysis is often decided judgmentally, taking into account both a statistical criterion and component interpretability. As part of this analysis, we examined four statistical criteria: 1) eigenvalue (λ_j) $\geq$ 1.0 before rotation (Rummel, 1970); 2) $\lambda_j \geq 1.0$ after rotation (Hopke, 1982); 3) unrotated λ_j greater than a critical value indicated by the Scree test (Rummel, 1970); and 4) unrotated λ_j greater than the 95 percent confidence interval of λ_j as estimated from a series of 50 randomly generated datasets having the same number of observations and variables as the data set being analyzed (Overland and Preisendorfer, 1982). In the last case,

the methodology employs the distribution of the λ_is derived from the 50 random data sets, and determines if the reported eigenvalue is significantly greater than those resulting from random noise alone. The results of these various tests are considered, as well as the interpretability of the resulting PCs, in order to arrive at the number of components retained for rotation in this work.

Following independent analyses of the meteorological and elemental particle data, interactions of the various source and meteorological components are investigated. This is accomplished by first computing the PC values (scores) on each sampling day. These scores are calculated from the original data by inverting Eq. (2) and are then correlated with the concurrent scores of the other PCs. Based on insights provided by the resultant correlation matrix of source and meteorological components, interpretations of the source components are clarified and the meteorological conditions associated with their impacts at the Watertown site identified.

3. Results

a. PCA of Watertown elemental particle data

The results of applying rotated PCA to the Watertown site fine elemental data set are presented in Table 3. The four statistical criteria considered for component retention provided conflicting guidance for this data set, ranging from three to five components (see Table 4). The five components actually retained, based upon both the results in Table 4 and component interpretability, account for 87 percent of the variance in the data set. The PCA structure presented in Table 3 contains the correlation r of each element with each rotated component. As previously suggested, these components are indeed interpretable as the underlying particle pollution sources affecting the Watertown site. The first component is identifiable as embodying the effects of soil particles upon the particle samples, since it is most correlated with the crustal elements (Si, Ca, Fe, Mn). The second component is noted as a motor vehicles component, based primarily upon its strong associations with Br and Pb, elements known to be enriched in leaded auto exhaust (e.g., Cooper and Watson, 1980). This is termed a motor vehicles component (not just leaded auto) in recognition of the fact that other motor vehicle particle impacts (including diesel truck and unleaded auto emissions) will be sampled at the monitor simultaneously with the leaded auto tracers. The third component is strongly correlated with both sulfur and selenium, and has been identified as being associated with coal combustion products (Thurston and Spengler, 1985). This initial assessment was based upon a number of considerations, primary of which is that coal combustion is the dominant anthropogenic source of Se emissions in the United States (Eimutis *et al.*, 1978). The fourth and fifth components are readily interpretable as particle source classes local to Boston. The residual oil component is highly correlated with V and Ni, two elements known to be enriched in residual fuels and their combustion products (Cooper and Watson, 1980). The final component is correlated with chlorine, an element indicative of the effects of salt aerosols at this site. These chloride aerosols are likely of seaspray and/or road salting origins in this northern coastal locale (Spengler and Thurston, 1983). Further details on the reasoning behind these source identifications (with estimates of their mass impacts and elemental compositions) have been presented in Thurston and Spengler (1985).

TABLE 3. Varimax rotated principal component analysis of Watertown fine mass elemental data (m = 332).

Element	Soil aerosols	Motor vehicles aerosols	Coal related aerosols	Residual oil aerosols	Salt aerosols
Si	0.97	0.05	−0.01	0.03	0.12
S	0.05	0.04	0.86	0.08	−0.11
Cl	0.26	0.06	−0.09	0.14	0.93
K	0.66	0.33	0.24	0.34	0.26
Ca	0.92	0.02	0.00	0.14	0.11
V	0.18	0.33	0.14	0.86	0.17
Mn	0.68	0.40	0.33	0.19	0.03
Fe	0.95	0.10	0.14	0.14	0.10
Ni	0.20	0.27	0.13	0.89	0.04
Zn	0.18	0.48	0.56	0.32	0.23
Se	0.11	0.10	0.88	0.08	0.01
Br	0.16	0.93	0.00	0.24	0.10
Pb	0.06	0.90	0.20	0.29	−0.05
			Eigenvalue		
	3.8	2.2	2.1	2.0	1.1

TABLE 4. Comparison of the results of various component retention criteria.

Method	Fine elemental data PCA	Coarse elemental data PCA	Meteorological data PCA
1. Unrotated $\lambda \geq 1.0$	4	4	6
2. Rotated $\lambda \geq 1.0$	5	6	10
3. Scree plot	5	6	5
4. λ Significance	3	3	5
PCs Actually Kept	5	6	5

The varimax rotated coarse mass elemental data PCA component structure is presented in Table 5. The component retention criteria recommendations (Table 4) ranged from three to six components for this data set. The six components retained account for over 95 percent of the total variance in the original fifteen variables. The interpretations of many of these components are similar to those for the fine data, as indicated by

TABLE 5. Varimax rotated principal component analysis of Watertown coarse mass elemental data (m = 332).

Element	Soil aerosols	Motor vehicle aerosols	Refuse burning aerosols	Residual oil aerosols	Salt aerosols	Sulfate aerosols
Al	0.97	0.06	0.06	0.14	−0.01	0.07
Si	0.98	0.04	0.06	0.12	0.00	0.05
S	0.17	0.19	0.10	0.23	0.07	0.93
Cl	0.00	0.04	0.00	0.13	0.99	0.06
K	0.97	0.05	0.04	0.11	0.05	0.10
Ca	0.94	0.05	0.05	0.14	0.02	0.05
Ti	0.97	0.04	0.05	0.16	0.00	0.04
V	0.25	0.20	0.07	0.89	0.10	0.13
Mn	0.97	0.04	0.07	0.11	0.00	0.03
Fe	0.97	0.07	0.09	0.17	0.00	0.08
Ni	0.35	0.27	0.24	0.77	0.11	0.20
Cu	0.00	0.06	0.95	0.00	0.01	0.00
Zn	0.21	0.16	0.87	0.24	−0.01	0.12
Br	0.05	0.97	0.09	0.15	0.03	0.09
Pb	0.08	0.96	0.12	0.20	0.02	0.11
Eigenvalue						
	6.8	2.0	1.8	1.7	1.0	1.0

the column headings. In this case, the PCA is also able to differentiate a coarse Cu–Zn particle component which has been identified as due to refuse incineration (Thurston and Spengler, 1985).

b. *PCA of Boston meteorological data*

The results of applying rotated PCA to the Boston meteorological data are shown in Table 6. As shown in Table 4, the eigenvector retention criteria indicated that from 5 to 10 components should be kept. The five meteorological components retained in this case accounted for 61 percent of the variance in the original 22 variables. This is a noticeably lower percentage of the total variance than was the case for either of the two elemental data sets. This result is likely due to the lower communalities among the original data in this case (ranging from 0.32 to 0.91) versus the trace element data (ranging from 0.73 to 0.99). While the reason for the higher communalities for the elemental data is not known with certainty, they may be caused by the fact that the pollution variables share a general rise and fall of concentrations as dispersion influences vary at the site considered.

The first meteorological component is noted as Eastern Seaboard (ES) transport, and is most strongly correlated with surface south winds (PERSS), rapidly decreasing surface pressure (− PTEND), and the presence of air mass advection up the Eastern Seaboard (PERCES). Examination of the weather maps for days with high scores for this component revealed that the component tends to reflect days in which a surface high pressure cell is well off the New England coastline

TABLE 6. Varimax rotated PCA application to Boston meteorological variables (m = 237).

Variable	Meteorological component: Eastern Seaboard transport	Moisture	Poor local dispersion	Season	Ohio Valley/Midwest transport
FLUX	−0.49	−0.48	0.26	0.29	−0.25
PRECIP	0.27	0.58	−0.36	0.04	0.07
SLPRESS	0.10	−0.09	0.64	−0.24	−0.20
PTEND	−0.70	−0.03	−0.30	0.06	−0.13
ATEMP	0.12	−0.05	0.14	0.90	0.17
AVGWSPD	−0.12	−0.23	−0.76	−0.11	−0.18
MVENTF	−0.42	−0.14	−0.55	−0.17	−0.08
AVENTF	0.02	−0.61	−0.26	0.04	0.08
RELHM	0.47	0.66	0.03	0.38	0.21
ABSHM	0.28	0.13	0.11	0.88	0.18
NSTBCL	0.01	−0.04	0.63	0.07	0.01
DSTBCL	0.26	0.09	−0.43	−0.26	0.02
PERNE	−0.03	0.69	−0.06	−0.05	0.07
PEREE	−0.06	0.56	0.31	0.24	−0.05
PERSE	0.11	0.30	0.48	0.20	−0.21
PERSS	0.79	−0.21	0.02	0.18	0.03
PERSW	0.35	−0.46	−0.03	0.20	0.52
PERWW	−0.50	−0.35	−0.23	−0.11	0.31
PERNW	−0.46	−0.20	−0.20	−0.30	−0.52
PERCES	0.65	0.15	−0.14	0.38	0.00
PERCOV	−0.18	−0.02	0.04	0.04	0.84
PERCEC	0.23	−0.11	0.17	−0.18	−0.76
Eigenvalue					
	3.1	2.8	2.7	2.4	2.2

and a surface low pressure cell is to the north of New England (with a trailing surface cold front in western New England). This synoptic regime results in strong winds in advance of the frontal passage. Wind trajectories indicate that the air masses tend to travel from the south-southwest (or along the east coast). However, frictional and thermal wind effects contribute to a more southerly flow at the surface, causing this component to be correlated with PERSS. This meteorological component appears to be strongly associated with air mass advection over the East Coast megalopolis toward New England.

The second component, moisture, represents the presence of synoptic conditions conducive to precipitation. Examination of the component pattern indicates that this regime is characteristically cloudy (− FLUX), high in relative humidity (RELHM), and experiences precipitation (PRECIP) and east (PERCEE) or northeast (PERCNE) winds. The afternoon ventilation factor is also negatively correlated with this component, as might be expected if there is little afternoon surface heating by the sun. This relationship between precipitation along the New England coast and synoptic northeast winds is most evident during North Atlantic Nor'easters, when there is a surface low pressure epicenter to the southeast of New England. Under northeast winds, the Watertown site will be affected by sources to the north of Boston, which include both industrial and power plant residual oil combustion sources.

The poor local dispersion component has a strong negative correlation with local windspeed (AVGWSPD), moderate positive correlations with nighttime inversions (NSTBCL), southeast winds (PERSE), and sea level pressure (SLPRESS), but a moderate negative correlation with daytime atmospheric stability (DSTBCL). This component embodies the effects of slow moving high pressure cells passing directly over the Boston area. The generally fair weather conditions associated with such a regime cause the noted increased frequency of nighttime inversions and daytime instability (vertical mixing). The moderate correlation with southeast winds appears to indicate the high frequency of sea breezes experienced in the Boston area during such high pressure, low synoptic wind conditions. Since Watertown is a western suburb of Boston, these conditions would tend to correlate with the impact of the Boston urban plume at the Watertown monitor.

The fourth component is most strongly correlated with average temperature (ATEMP) and mean absolute humidity (ABSHM). Since water's saturation vapor pressure is a direct function of temperature, it is not surprising that absolute humidity and temperature vary together. In the Boston area, this component is strongly seasonal in nature (i.e., high in summer and low in winter). Although a measure of ambient oxidants is not included in this data set, a previous factor analysis of a data set including meteorological and air pollution variables for Boston has shown that photochemical oxidants, which tend to be highest in the summer months, are highly correlated with this component (Thurston and Spengler, 1980). Overall, this component represents the effects of seasonal fluctuations on the data.

The final component is positively correlated with conditions conducive for the transport of air masses from the Ohio Valley/Midwest sector (PERCOV), and negatively correlated with conditions conducive to air mass transport from eastern Canada (PERCEC). It is not correlated with any specific local meteorological measures, save moderate positive correlations with southwest and west surface winds, and a negative correlation with northwest surface winds. Overall, this component appears to reflect the fact that Ohio Valley/Midwest transport and eastern Canada transport trajectories are associated with distinct (and mutually exclusive) meteorological regimes.

c. *Correlation analyses of elemental and meteorological components*

The correlation matrix in Table 7 presents the relationships among the derived source and meteorological components. The PCA has distilled two correlation matrices of 13 and 15 elements for the fine and coarse mass data sets, respectively, with 22 meteorological variables (or 616 paired correlations) down to a more manageable five by eleven matrix (55 paired correlations) among 16 physically interpretable components. This matrix can now be analyzed to derive the interactions of the various meteorological phenomena (represented by the five meteorological components) with each of the eleven source components.

Before examining the correlation matrix, it is important to make two comments of a statistical nature. First, although the correlation matrix has been distilled substantially, there are still 55 correlations tested. If $p = 0.05$ were used as the criterion for statistical significance for each individual test, then it could be expected that two correlations would be indicated as statistically significant by chance, even if there were no true correlation among the variables considered. Thus, the Bonferonni inequality is invoked, which states that (Harris, 1975):

$$p_{\text{overall}} \leq \sum_i p_i, \tag{3}$$

where p_i is the Type I error for each test (i.e. error pertaining to the probability of falsely rejecting the null hypothesis if it is true). Thus, in order that the p_{overall} be approximately 0.05, the individual test of significance applied to each of 55 paired correlations is $p = 0.001$. Second, it should be remembered that the principal components are not direct measures of the processes they represent, but are instead derived indicators. As a result, it has been found that their scores generally do not achieve correlations as large as direct measures would (Harris, 1975). This analysis examines

TABLE 7. Paired correlations between the Watertown, MA source and meteorological components (m = 237).

Source component	Meteorological component: Eastern Seaboard transport	Moisture	Poor local dispersion	Season	Ohio Valley/Midwest transport
Fine mass PCs					
Soil	−0.12	−0.20	−0.01	−0.26*	0.04
Motor vehicles	0.06	0.19	0.42*	−0.22*	0.08
Coal related	0.27*	−0.18	0.14	0.25*	0.59*
Residual oil	0.15	0.22*	0.37*	−0.26*	−0.03
Salt	0.00	0.16	−0.16	−0.18	0.04
Coarse mass PCs					
Soil	−0.13	−0.25*	0.12	0.39*	0.02
Motor vehicles	0.03	0.11	0.23*	−0.01	−0.02
Refuse burning	−0.05	−0.04	0.23*	0.11	−0.01
Residual oil	0.13	0.12	0.27*	−0.24*	0.13
Salt	0.05	0.10	−0.02	−0.19	−0.02
Sulfate	0.23*	0.13	0.02	0.44*	0.31*

* Statistically significant at $p = 0.001$ (i.e., $P_{\text{overall}} \leq 0.055$).

correlations of the derived meteorological component scores with the derived source component scores. Thus, in the following discussions, correlations of the order of ±0.5 are considered to be relatively strong, while those from approximately ±0.25 to 0.50 should be viewed as relatively moderate.

Air mass transport along the Eastern Seaboard to the Boston area (ES Transport) is not strongly correlated with any of the particle pollution components identified in Table 7. There is, however, a moderate correlation of ES transport with fine coal combustion related aerosols, as well as with the coarse sulfates component. These correlations are not surprising, given the concentration of SO_2 emissions within the East Coast megalopolis between Washington, D.C. and Boston (Clark, 1980; Benkovitz, 1982).

Moisture effects are moderately (and negatively) correlated with soil impacts, indicating the effects of precipitation in reducing fugitive dust resuspension. It seems doubtful, however, that the moderate correlation between moisture and residual oil impacts is due to the effects of high relative humidity or precipitation. Instead, since this component is moderately correlated with northeast winds, it seems probable that the correlation is a manifestation of the impact of residual oil combustion products emitted from sources to the north of Boston during winds from the northeast quadrant.

Reduced dispersion of pollutants (Poor Local Dispersion) has its greatest correlation with local combustion source impacts, being most correlated with fine mass motor vehicle impacts, but also being moderately correlated with coarse and fine mass residual oil combustion, coarse mass motor vehicle emission, and refuse incineration aerosol contributions. This meteorological component's strong influence on local combustion source impacts at the Watertown site is likely due to a combination of the influences of poor emission dilution and the fact that sea breezes (southeast winds) tend to be associated with this meteorological regime, causing mesoscale advection of central Boston's pollution toward the Watertown site. It is interesting to note that all significant correlations with poor dispersion are positive, confirming the previously hypothesized shared rise and fall of many elemental concentrations with local dispersion.

Seasonal effects show the greatest number of significant correlations with the particle pollution components. This component is negatively related to local combustion sources (fine and coarse residual oil and motor vehicle impacts), which appears indicative of higher emissions of these sources in the winter months (perhaps due to greater heating oil use and higher gram per mile emissions during cold startups of autos). Conversely, season is positively correlated with fine coal combustion related aerosols, sulfates, and coarse soil impacts. These correlations indicate added sulfate formation in the photochemically active warm months, combined with a lack of snow cover to inhibit soil particle generation. One anomalous moderate correlation with season is the fine soil component, which (unlike the coarse soil component) is negatively correlated with season, indicating the impacts to be higher in winter than in summer. The reason for this relation is unclear, and it may draw into question the component's interpretation as fugitive soil related. In general, however, these correlations confirm our interpretations of the various components, with the combustion source impacts being highest in winter and the sulfur related and coarse soil components being highest in the summer months.

The highest value reported in Table 7 is the correlation between air mass transport across the Ohio Valley/Midwest sector (OV transport) and the coal related aerosol component. This correlation is much stronger than was the case for coal and ES transport, indicating the Ohio Valley/Midwest sector to be the primary (but not sole) source region for air masses containing this pollution mix. Persistent southwest wind flows, such as those indicated for the OV Transport component, typically occur on the northwest side of a surface high pressure cell and/or in the warm sector of a surface low pressure cell. These conditions have been shown to be conducive to the long distance transport of fine particulate aerosols in other studies (e.g., Lavery *et al.*, 1979 and Tong and Batchelder, 1978). This correlation between the transport of air masses across the Ohio Valley/Midwest region and the values of the fine aerosol component identified as coal combustion-related aerosols (and their associated sulfates), irrespective of local dispersion conditions, is the clearest pollution source–meteorological component association identified by this statistical analysis.

The only other significant correlation noted with OV transport in Table 7 is for the coarse sulfate component, which shows a moderate relationship in the same direction as the coal combustion related component. However, it seems very unlikely that coarse sulfur particles would undergo long distance transport. Given that coarse sulfur levels are roughly one-tenth the fine sulfur levels at this site, on average, and that the virtual impactor allows a fraction of fine particle mass to cross over onto the coarse filter (e.g., see McFarland *et al.*, 1979), a plausible explanation may be that a portion of the "coarse" sulfate detected is due to the crossover of fine particle sulfates onto the coarse filter.

In order to further investigate the above noted OV transport–coal related aerosol relationship, the individual air mass trajectories for those days having the 10 highest and 10 lowest coal related impacts were examined. Figure 2 displays the midday air mass trajectories for these 20 days. These trajectories are the estimated air mass paths ending in Boston on the days of interest (1:00 p.m. eastern standard time, except in one case when wind data were missing, and the 7:00 a.m. trajectory was employed). As discussed earlier, these trajectories were derived from observed mixing layer average winds throughout the United States using the ARL-ATAD model. It is seen that the highest impact days have trajectories enveloping the industrialized United States, while the lowest impact days' trajectories primarily pass over eastern Canada. These results concur with work by Parekh and Husain (1981) which showed a similar directionality of an S–Se–As component they identified as coal related, and with the work of Shaw and Paur (1983), which indicated Ohio Valley aerosols to be highly enriched in Se. This coal-related component is strongly associated with conditions conducive to the transport of polluted air masses into the Boston area.

Finally, it should be mentioned that a combined trace element and meteorological variable data set was also analyzed via a single PCA for comparison with

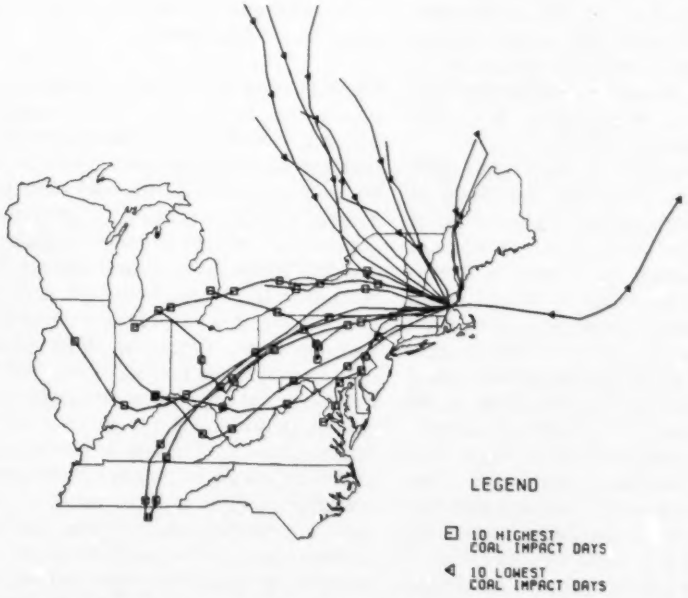

FIG. 2. Air mass trajectories arriving midday in Boston on the ten highest and ten lowest estimated coal related aerosol impact days. (Symbols are separated by 12 h travel time.)

the above results. The resulting source–meteorological components, presented elsewhere (Thurston, 1983), tend to confirm the major relationships noted above, while resulting in fewer components having higher shared variances among elemental and meteorological variables. However, since rotated PCA tends to result in each variable being strongly correlated with only one of the components, it was found that the weaker meteorological-source relationships noted above were not revealed by the combined dataset PCA. For this reason, the separate analysis of the meteorological and elemental data with subsequent correlation analyses appears to be the preferable approach in analyzing the source–meteorology interactions.

4. Conclusions

The application of varimax rotated principal component analysis to separate elemental and meteorological data sets provides new composite variables (components) which are interpretable, respectively, as particulate source classes and meteorological regimes influencing the site considered. In the process of conducting the PC rotations, we have considered four different component retention criteria. The number of components actually retained in this work was based upon both the guidance indicated by the various criteria considered and upon our assessments of whether the components were interpretable as plausible influences on the data. It was found that the number of components decided upon was usually more than indicated by $\lambda_j \geq 1.0$ before rotation, but that all components kept had $\lambda_j \geq 1.0$ after rotation. The eigenvalue significance test was the most restrictive criterion, always indicating fewer components than the other methods considered. It became apparent that this technique is similar to, but somewhat more restrictive than, the $\lambda_j \geq 1.0$ before rotation approach. Functionally, it required that the middle eigenvalue (e.g., for the eighth of 15 components) be greater than approximately 1.0, with the first half of the eigenvalues needing to be higher still and the second half needing to be higher than a (declining) critical eigenvalue less than 1.0. For these specific data sets, the Scree test for PC retention was found to consistently result in the most interpretable rotated components. At this point, these indications of the usefulness of the various component retention criteria are relevant to the data sets considered, and might not be generalizable to other elemental or meteorological data sets.

A pairwise correlation analysis of the derived source and meteorological component scores has provided insights into the role of meteorological influences in determining the variability of identified particle source impacts, and has tended to confirm previous interpretations of the source components (Thurston and Spengler, 1985). Of the source classes identified via the elemental data PCAs, the local combustion sources (residual oil burning and motor vehicle emissions) were found to correlate most strongly with a poor local dispersion component in the meteorological data. A coarse soil component (related to the crustal elements Al, Si, Fe, Ca, Ti, and Mn) was found to be lowest during the winter (snow cover) season and during precipitation, both of which are known to reduce fugitive dust generation. Finally, the strongest relationship reported in this analysis involved a fine particle Se and S component, which had been identified as transported coal combustion related aerosols (Thurston and Spengler, 1985). This component's interpretation was confirmed by the correlation analysis, as it was found to be highly correlated with the presence of air mass transport over the Ohio Valley/Midwest region to Boston, irrespective of local dispersion conditions. These results indicate that both local and transported air pollution contribute to the inhalable particle exposures in metropolitan Boston, with the sulfate portion being highest during polluted air mass transport into the Boston area.

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